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Multiple closed loop recycling of carbon fibre composites with the HiPerDiF (High Performance Discontinuous Fibre) method

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A B S T R A C T
The aim of this article is to apply the concept of Circular Economy, where end-of-life products and production wastes are recycled into reusable materials, to carbon fibre reinforced plastics. This not only reduces the amount of material disposed into landfills, but also enables manufacturers to achieve significant savings. While current research focuses on the performance of recycled carbon fibre reinforced composites after one recycling process, this paper aims to investigate the performance of composites remanufactured from short carbon fibres that have undergone multiple recycling loops with the High Performance Discontinuous Fibre (HiPerDiF) method. The HiPerDiF method enables the production of aligned short fibre composites with exceptional mechanical properties. In addition, using short fibres makes the composite material intrinsically easy to recycle. Short virgin carbon fibres underwent two loops of fibre reclamation and remanufacturing. A correlation between the composites’ mechanical properties and the nature of the fibres, i.e. reduction in fibre lengths, as well as the residual matrix accumulation from the reclaiming process over a number of recycling loops, was established.

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1. Introduction
Circular Economy is defined as an economy that is restorative and regenerative by design, and which aims to keep products, components and materials at their highest utility and value at all times [1]. Its application to the recycling of carbon fibre reinforced plastics (CFRPs) has been a subject undergoing intense study both in the academic and industrial fields. In the academic field, various fibre reclaiming methods and remanufacturing processes have been proposed by researchers [2]. On the commercial side, aircraft manufacturers, in partnership with external organisations, have planned various projects to recycle CFRPs. At Airbus Composite Technology Centre, the development of a recycled carbon fibre veil prepreg to replace current glass fibre products used in aircraft interior applications is being explored [3]. Boeing has manufactured small access doors on the underside of the airplane’s wings using carbon fibre wastes from the production of 787 Dreamliner trailing edges [4]. According to the UK Composites Strategy [5], the government recognises the need to improve recycling processes and develop added-value applications for recycled composites. In United States, a bill was passed in the Senate to study ‘the technology of recycled carbon fibre and production waste carbon fibre’ as well as ‘the potential lifecycle energy savings and economic impact of recycled carbon fibre’ [6].

The current method of handling CFRP at their end-of-life has a negative impact on the environment as they are typically disposed of in landfills. With some 6000–8000 commercial planes expected to reach end-of-life dismantlement by 2030 [7], the environmental impact of the increasing use of CFRP will worsen. In an effort to mitigate this, legislation has been put in place. This includes among others, the landfill directive and tax as well as the incineration of waste directive [8]. The use of recycled carbon fibres, not only alleviates the environmental impact of disposing CFRPs into landfills, but also helps manufacturers achieve significant savings. Currently, carbon fibres used in the aerospace industry are of the highest grade and cost between $5 and $50/lb [9] as virgin materials. From the cost breakdown shown in Fig. 1, the high cost of virgin carbon fibres is mainly driven by its manufacturing cost.

Boeing estimated that carbon fibres can be recycled at approximately 70 per cent of the cost to produce virgin fibres ($8/lb to $12/lb vs. $15/lb to $30/lb) while using less than 5 per cent of the electricity required (1.3–4.5 kWh/lb vs. 25–75 kWh/lb) [11].

Pimenta and Pinho [12] present a review of the current status and an outlook of CFRP recycling operations, focusing on state-of-the-art fibre reclamation and remanufacturing processes. The industrial process of recycling scrap CFRP usually begins with the sorting and shredding of the scrap composites into smaller parts. These parts subsequently undergo fibre reclamation process to remove
the resin from the scrap composites. The most commonly used fibre reclamation processes are pyrolysis [13,14], oxidation in a fluidised bed [15] and chemical recycling [16,17]. Although the fibre reclamation process differs from one company to another, i.e. pyrolysis for ELG Carbon Fibre Ltd. [13] and wet chemical breakdown in Adherent Technologies, Inc. [18], the main aim of the process is the same: to remove as much resin and sizing agent as possible from the fibre surfaces while minimising damage to the fibres.

After the reclamation process, the fibres are fluffy and highly entangled. Subsequently, the fibres undergo various post-reclaiming processes according to the desired type of product form, e.g. wet-laid manufacturing to produce non-woven mats [19,20], milling or chopping [12]. Currently, the main products in the market are non-woven mats as well as chopped and milled fibres from comminution processes [12]. The reclaimed carbon fibres can be used in conventional methods such as compression moulding for non-woven mats and injection moulding for pellets. However, these methods produce composites that have limited performance as they are typically low in fibre content [12] and the control of fibre orientation is difficult [21]. Due to the nature of the products, i.e. milled and short random carbon fibres, they are typically employed in non-structural applications, this fails to maximise the value of recycled carbon fibres.

In order to obtain recycled composites that have properties comparable to virgin CFRPs in high performance applications, high fibre volume fraction (40% and above) is necessary. One way of achieving this is to produce recycled CFRPs that contain highly aligned carbon fibres. A study by Guell and Graham [22] showed that highly aligned short fibre composites have 90% higher stiffness and an increase of 100% of the tensile strength in the alignment direction compared to randomly oriented fibre composites. When highly aligned short fibre composites were compared to continuous fibre composites, Flemming et al. [23] demonstrated that 94% of the tensile stiffness and 80% of the strength of the continuous fibre composites were retained by the highly aligned short fibre composites. Hence, the potential of obtaining short fibre reinforced composites that have mechanical properties comparable to continuous fibre composites lies in fibre alignment. This was also pointed out by Carberry, program manager for Boeing’s Airplane and Composite Recycling, “The key to unlocking the manufacturing future for recycled fibre beyond injection moulded applications is aligning the fibres.” [11].

The various methods of aligning carbon fibres can be divided into two main categories: dry and wet fibre alignment. Dry fibre alignment methods include the use of magnetic or electric field while the latter makes use of hydrodynamic forces. These methods were evaluated and their results consolidated in a study by Smith [24]. It was found that in the magnetic field method by Yamashita et al. [25], fibre alignment became difficult to control when the fibre volume fraction reached 5%. The electric field method developed by Kim et al. [26] gave more promising results with approximately more than 50% of the fibres aligned to within 5°. However, the inability of these methods to produce composites with sufficient level of fibre alignment and high fibre volume fraction limited their use in high performance applications. Wet fibre alignment methods have been seen as a solution to these problems. Hydrodynamic methods typically involve suspending fibres in a liquid and accelerating the mixture through a converging nozzle [27], the alignment is achieved by subjecting the fibres to differences in the fluid velocity. Hence, it is dependent on the characteristics of the fibres, i.e. aspect ratio and stiffness, as well as the concentration of fibres in the liquid medium [28]. Another important parameter that determines the fibre alignment is the viscosity of the fluid. Bagg et al. [29] discovered that glycerine was most suitable as a medium to facilitate the achievement of maximum fibre alignment in a short fibre reinforced composite. The process was also adopted by Kacir et al. [30]. Both results presented high degrees of fibre alignment. The former showed that around 80–90% of the fibres were oriented to within 10° of a common axis and in the latter study, over 90% of the fibres were found in the angular range of ±15°. Therefore the wet fibre alignment methods have been tried as remanufacturing technique for reclaimed carbon fibres by several researchers. Wong et al. [31] proposed a centrifugal alignment rig, which uses a dispersion of fibres in a viscous media accelerated through a convergent nozzle installed in a rotating drum. An alignment level of 90% was obtained using 5 mm reclaimed carbon fibres. The same authors worked on a hydrodynamic spinning process of a viscous fibre suspension. However, despite achieving highly aligned fibres in the aforementioned studies, these methods are not widely adopted as they require the washing of the viscous carrier media and drying after remanufacturing, this limits the productivity to around 4 kg/h [27]. A modified papermaking technique was also applied to reclaimed fibres by Pickering [32], Turner et al. [33] and Warrior et al. [34] reaching 80% of the theoretical alignment value and a fibre volume of 45% with a moulding pressure of 100 bar. However, the fibre alignment level was not sufficient to reach high fibre volume fraction under low manufacturing pressure. A recent study by Yu et al. [35] addressed this issue by introducing a novel method, named High Performance Discontinuous Fibre (HiPerDiF). This method uses the momentum change of a fibre suspension in a low viscosity medium to align discontinuous fibres. Results showed good alignment of the discontinuous fibres with 67% of the fibres aligned within the range of ±3°. Higher mechanical properties compared to composites from conventional fibre alignment methods were obtained. The use of water instead of glycerine as a fluid medium also means that reduction in remanufacturing time and costs can be attained.
Current research has shown considerable success in optimising the recycling processes to obtain recycled CFRPs for high performance applications. However, studies conducted thus far focus on mechanical properties of recycled composites which were remanufactured with fibres that have undergone only one recycling process. This does not maximise the value of reclaimed carbon fibres which may be capable of being recycled again. In this paper, a closed loop of recycling process is defined by combining a suitable fibre reclaiming process and a remanufacturing methodology to obtain a high performance material. Pyrolysis has proven to be the most effective fibre reclaiming process to remove resin from the fibres while retaining the fibres’ mechanical properties. For the remanufacturing process, the HiPerDiF method has shown the greatest potential to efficiently align short carbon fibres to a high degree without the use of a viscous medium. In addition, this method produces composites that can be recycled easily: chopped virgin fibres are compatible with the reclaimed fibre remanufacturing process, which eliminates the need for post reclaiming comminution. Therefore, using the two selected processes in a closed recycling loop, this paper aims to investigate the performance of recycled composites made from fibres that have undergone multiple recycling closed loops. This represents a potential move towards a Circular Economy for businesses in the CFRP industry.

2. Experimental

2.1. Methodology

An overview of the experimental work is shown in Fig. 2. Virgin carbon fibre (vCF) specimens are manufactured with the HiPerDiF method and tested to evaluate their mechanical properties. The specimens are then subjected to pyrolysis and new specimens are remanufactured with the reclaimed carbon fibres (rCF) and tested. This process is repeated two times.

2.2. Materials

Prepregs were manufactured with the HiPerDiF method using 3 mm carbon fibres (HTS40, Toho Tenax) and epoxy resin film (MTM49-3, Cytec). The fibre properties provided by the manufacturer are shown in Table 1 [Toho fibre].

<table>
<thead>
<tr>
<th>Table 1 Carbon fibre properties [Toho fibre].</th>
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<tbody>
<tr>
<td>Toho Tenax HTS40</td>
</tr>
<tr>
<td>Diameter [μm]</td>
</tr>
<tr>
<td>Length [mm]</td>
</tr>
<tr>
<td>Density [g/cm³]</td>
</tr>
<tr>
<td>Young’s modulus [GPa]</td>
</tr>
<tr>
<td>Failure σ₁₁ [MPa]</td>
</tr>
<tr>
<td>Failure ε₁₁ [%]</td>
</tr>
</tbody>
</table>

(Meyer et al. [36].) The fibre properties provided by the manufacturer are shown in Table 1 [Toho fibre].

2.3. HiPerDiF method and sample preparation

The HiPerDiF method (steps 2 and 7 in Fig. 2) works with a low concentration suspension of carbon fibres in water. The suspension is accelerated by a peristaltic pump through nozzles that are directed towards the orientation head placed above a perforated conveyor belt with a suction system underneath. The orientation head is made of a series of parallel plates spaced by a controllable gap. When the fibres hit the plates, their orientation changes transversely to the water jet direction, provided that the gap between the parallel plates is a maximum of 1/3 of the fibre length. As they fall on the conveyor belt, the fibres realign themselves to the movement direction of the belt to produce the preform. The water is removed by vacuum suction, as shown in Fig. 3.

The preform is then dried from any remaining water using infrared radiation. The aligned fibre preform is coupled with an epoxy resin film and partially impregnated by applying heat and pressure. The prepregs were laminated in a unidirectional lay-up in a semi-closed mould, and cured by vacuum bag moulding in an autoclave at a pressure of 7 bar, and temperature of 135 °C for 90 min to produce the specimens (step 3 in Fig. 2).

2.4. Fibre reclaiming process – Resin burning

The fibre reclamation process, i.e. resin burning (step 5 in Fig. 2), was determined based on the study by Meyer et al. [36].

Fig. 2. Overview of the closed recycling loop.
When pyrolysis was conducted in atmospheric air, it was found that at a temperature of about 600 °C, there was complete removal of resin. But at temperatures of 650 °C and above, there was further weight loss which implied that oxidation of the fibres and a decrease in fibre properties might have occurred. Hence, in the resin burning process, a more conservative temperature of 500 °C was chosen. This temperature was selected to account for differences in the type and amount of carbon fibres used in this experiment and in Meyer’s study, as well as ensure minimal risk of damaging the fibres. Fig. 4 shows that compared to other temperatures, operating at a temperature of 500 °C in air atmosphere resulted in the decomposition of almost all of the former epoxy with minimal further weight loss of the fibres and independent of the dwelling time.

The selected fibre reclamation process was therefore the pyrolysis in atmospheric air for 5 h at 500 °C to ensure that the temperature was uniform throughout the furnace and all the fibres would be treated. This also enabled the removal of the glass fibre end-tabs and the rubber toughened epoxy adhesive, which was used to bond the end-tabs to the specimens. This process also allowed the fibre volume fraction of the specimens to be obtained according to ASTM D2584-11 [37]. To remove any contamination, the reclaimed fibres were washed in an ultrasonicator for 5 min in a solution of water and 20% acetone by volume. The washed fibres were then dried in an oven for 1 h at 100 °C.

2.5. Fibre length measurement and SEM imaging

The fibre length of each manufactured specimen set was measured before each HiPerDiF manufacturing step (steps 1 and 6 in Fig. 2). Changes in the fibre surface of each specimen set were also observed using the Scanning Electron Microscope.

2.6. Mechanical Testing

Referring to ASTM D3039/D3039M-14 [38], tensile tests were performed on the specimens using a servo-electric tensile test machine (step 4 in Fig. 2). GFRP end-tabs were bonded to the both ends of each specimen with rubber toughened epoxy adhesive (Araldite 2014). White dots were sprayed onto the surface of each specimen to measure the strain with a video-gauge system. Fig. 5 presents a schematic of the specimen.

The specimens were loaded at a constant cross-head displacement speed of 1 mm/min and the load was recorded with a 10 kN load cell.

The fibre reclamation and remanufacturing process was repeated twice to achieve two recycling loops as illustrated in Fig. 2. The specifications of the specimens according to the recycling loops are shown in Table 2.

The reduction in the number of specimens after the second recycling loop was due to the loss of fibres during the reclamation and remanufacturing process.

3. Results and discussion

Representative stress–strain curves obtained by the tensile test of step 4 of each recycling loop are shown in Fig. 6.

All the three sets of specimens showed a linear-elastic tensile behaviour and brittle failure. A decrease in stiffness and failure properties can be observed.

In order to compare the results, the elastic modulus and failure strength results for the rCF1 and rCF2 specimens were normalised against the fibre volume fraction of the vCF specimen. The measured fibre volume fractions of the specimens are listed in Table 3.

The vCF composite shows a lower fibre volume fraction when compared to the one obtained by Yu et al. in [39], this is caused by a lower areal weight of the manufactured preforms and a higher resin film areal weight.

The fibre length distribution of each manufactured specimen set, obtained by measuring the length of a random sample of 200 fibres with an optical microscope and dedicated software, is shown in Fig. 7.

Observing Table 3, the drop in fibre volume fraction from vCF and rCF1 can be explained by a reduction of fibre alignment level caused by the shortening of the fibres between the manufacturing of vCF and rCF1 specimens. The gap between the parallel plates of the alignment head has been kept constant for the three remanufacturing process. This, combined with a significant reduction of the longer fibres between vCF and rCF1, caused a sensible...
reduction in the fibre alignment level and the formation of resin rich regions, as observed by Yu et al. in [35]. This, on one hand, causes a reduction of the stiffness and, on the other hand, a reduction of the fibre volume fraction, as a higher level of alignment means that the fibres are more tightly packed, as observed by Wong in [31]. Moreover, some of the shorter fibres are lost during the wet alignment process as they are not withheld by the perforated mesh belt, reducing further the fibre volume fractions. The fibre length reduction can be mainly attributable to damage during the mechanical test, the harsh reclaiming parameters and the post-reclamation washing process. The amount of the fibres in the length range between 1.6 and 2.4 mm by 66% after the first reclamation process increased only by 21% after the second reclamation process, leading to a similar level of fibre alignment and therefore of fibre volume fractions. The drop in fibre volume fraction can be addressed by grading and fractioning the fibres with techniques adopted from the paper recycling processes [40,41].

From the tensile tests, the modulus, failure strength as well as the failure strain of the specimens were determined and tabulated, as shown in Table 4. The measured elastic moduli and failure stress have also been normalised to a common fibre volume fraction of 38%.

The results presented in Table 4 are visualised in Figs. 8 and 9. The drop in measured material stiffness between vCF and rCF1 can be explained considering the reduction in fibre volume fraction. As a matter of fact the normalised elastic moduli between

<table>
<thead>
<tr>
<th>Fibre volume fraction</th>
<th>Mean [%]</th>
<th>CV [%]</th>
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<tbody>
<tr>
<td>Virgin carbon fibre composite (vCF)</td>
<td>37.7</td>
<td>2.4</td>
</tr>
<tr>
<td>Recycled composite 1 (rCF1)</td>
<td>28.3</td>
<td>0.64</td>
</tr>
<tr>
<td>Recycled composite 2 (rCF2)</td>
<td>28.5</td>
<td>0.18</td>
</tr>
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</table>

Table 3
Fibre volume fraction of composites as function of recycling loops.

![Fig. 6. Representative stress–strain curves.](image)

![Fig. 7. Fibre length distribution as function of recycling loops.](image)

![Fig. 8. Measured and normalised tensile modulus of composites as function of recycling loops.](image)

![Fig. 9. Normalised failure stress and strain of composites as function of recycling loops.](image)

<table>
<thead>
<tr>
<th>Elastic modulus measured</th>
<th>Mean [GPa]</th>
<th>CV [%]</th>
<th>Elastic modulus normalised</th>
<th>Mean [GPa]</th>
<th>CV [%]</th>
<th>Failure stress normalised</th>
<th>Mean [MPa]</th>
<th>CV [%]</th>
<th>Failure strain</th>
<th>Mean [%]</th>
<th>CV [%]</th>
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<tr>
<td>Virgin carbon fibre composite (vCF)</td>
<td>65.9</td>
<td>7.1</td>
<td>65.9</td>
<td>7.1</td>
<td>731</td>
<td>18.6</td>
<td>1.11</td>
<td>16.2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Recycled composite 1 (rCF1)</td>
<td>52.0</td>
<td>5.4</td>
<td>69.3</td>
<td>7.2</td>
<td>614</td>
<td>13.1</td>
<td>0.89</td>
<td>8.1</td>
<td></td>
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<tr>
<td>Recycled composite 2 (rCF2)</td>
<td>40.4</td>
<td>5.5</td>
<td>53.3</td>
<td>7.3</td>
<td>279</td>
<td>53.6</td>
<td>0.52</td>
<td>12.4</td>
<td></td>
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</tbody>
</table>

Table 4
Experimental results as function of recycling loops.
vCF and rCF1 is constant: this shows that the reclamation process was successful in achieving the requirement of minimising fibre stiffness degradation during the recycling process and maximising the performance of recycled fibres through the remanufacturing stage. However, even if no sensible fibre volume fraction change was recorded between rCF1 and rCF2, the drop in measured stiffness is accompanied by a reduction in normalised stiffness, see Fig. 8. This can be explained by the combination of three factors: a significant loss of the longer fibres after the second recycling loop, observable in Fig. 7; a reduction in the fibre stiffness, caused by degradation during the reclaiming process; a reduced adhesion between the fibre and matrix caused by residues build up on the fibre surface, as shown below.

As widely recognised in literature, the pyrolytic reclamation process reduces the fibre strength [42], this is confirmed by the trends of normalised failure stress and strain in Fig. 9.

Comparing the SEM images of fibres used to manufacture the vCF specimen (Fig. 10(a)), rCF (Fig. 10(b)) and rCF2 (Figs. 10(c)) specimens, it can be seen that there was an increasing amount of residual matrix left on the fibres after each round of resin burning process. vCF generally have a clean surface while rCF1 have some epoxy matrix attached to them. The most amount of epoxy matrix left on the fibre surface was observed on rCF2.

Results from the SEM images correlate to results from the tensile tests. vCF had the highest mechanical properties as the fibres have a clean surface. The first resin burning process was successful in removing most of the resin from the fibre, thus allowing adhesion between the fibre and matrix. However, the increasing amount of residual matrix on the fibres used to manufacture the rCF2 inhibited the adhesion of the new matrix to the fibres during remanufacturing. As a result, a large decline in failure strength was observed.

The failure strain results, shown in Fig. 7, present a similar trend to the failure strength results with the exception of the reduced coefficient of variation of rCF2. With increasing recycling loops, the strain to failure decreased. As mentioned earlier, the multiple recycling processes caused the reduction of fibre length, which reduces the efficiency of the load transfer from the matrix to the fibres. The reduction in strength over subsequent cycles can be therefore attributed to the combination of the reduction in strength caused by the pyrolytic reclamation process and the build-up of residual resin over the reclaimed fibre surface.

![Fig. 10. SEM images: (a) Virgin carbon fibres. (b) Fibres after the first recycling loop. (c) Fibres after the second recycling loop.](image-url)
4. Conclusions

Results from the experimental work have proven that with the chosen fibre reclaiming and remanufacturing processes, maximum retention of the mechanical properties of virgin carbon fibre composites after the first round of recycling can be obtained. However, after the second round of recycling, recycled composite specimens showed a decline in the stiffness as well as failure strength and strain. Further tests, i.e. fibre length measurements and SEM imaging, showed that the decrease in the mechanical properties is due to the shortening of the fibres and to the accumulation of residual matrix on the fibre surface, which inhibited interfacial adhesion between the fibres and the matrix. Optimising the fibre reclaiming process and avoiding fibre damage will help to retain higher mechanical performance over consecutive recycling loops.

In order to maximise the use of recycled carbon fibres, future work will aim to improve the mechanical properties of recycled composites by addressing these issues:

1. Optimisation of the fibre reclaiming process is needed for multiple recycling loops in order to prevent the accumulation of matrix on the fibre surface.
2. A sizing agent should be introduced into the remanufacturing process to minimise damage to the fibres.
3. Fibre grading and fractioning should be introduced to eliminate the shorter fibres and maintain high stiffness and fibre volume fraction in the recycled composites.
4. Hybridisation with virgin carbon fibre could also be considered to replenish the discharged reclaimed fibres.

Acknowledgements

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Supporting data are available, subject to a non-disclosure agreement. Please contact the corresponding author in the first instance.

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